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STUDY OF ELASTIC PROPERTIES NEAR A
NEMATIC-SMECTIC A TRANSITION

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STUDY OF ELASTIC PROPERTIES NEAR A
NEMATIC-SMECTIC A TRANSITION

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ABSTRACT

Measurements of the splay and bend elastic constants of octyl-cyano biphenyl near the nematic-smectic A transition are presented. The pretransitional increase of K_3 obeys the relation $\delta K_3 \sim (T-T_{SN})^{-\nu}$, with $\nu = 1$.

АННОТАЦИЯ

Были измерены константы упругости деформации поперечного и продольного изгиба на октил-циано-бифениле. Вблизи нематико-смектического перехода K_3 увеличивается по закону $\delta K_3 \sim (T-T_{SN})^{-\nu}$, где $\nu = 1$.

KIVONAT

Oktil-ciano bifenil feszítési és hajlítási rugalmas állandóját mértük a nematikus-szmektikus A átmenet közelében. K_3 növekedése a fázisátmenet közelében a $\delta K_3 \sim (T-T_{SN})^{-\nu}$, $\nu = 1$ összefüggésnek tesz eleget.

1. Introduction

In the last few years a large number of substance have been syntetised, which exhibit second order or weakly first order nematic-smectic A transition. Near transitions of these types pretransitional phenomena are observed in the nematic range, which are due to the presence of small clusters in which the smectic order has already developed.

A typical pretransitional effect is the temperature dependence of the Frank-elastic constants near the transition. While the splay elastic constant (K_1) does not show any sharp change, the twist (K_2) and bend (K_3) elastic constants increase rapidly as the transition temperature is approached. This fact is explained by the structure of the smectic A phase; in this phase only splay is allowed, twist and bend are forbidden. The gradual appearance of regions with smectic order in the nematic phase leads to the increase of K_2 and K_3 .

A number of measurements of the bend and twist elastic constants near nematic-smectic A transitions have been reported recently. The data are usually interpreted by assuming that the increment of K_3 or K_2 is proportional to some power of $T - T_{SN}$ (T_{SN} transition temperature):

$$\delta K_3 \sim (T - T_{SN})^{-\nu} \quad /1/$$

Different authors have found different values for ν ; e.g. P.Cladis on CBOOA $\nu = 0.52$ [1]; Pindak et. al. also on CBOOA $\nu = 0.68$ [2]; Cheung and Meyer on p-butoxybenzylidene-p'- β methylbutylaniline, which has a weakly first order nematic-smectic A transition, have found $\nu = 1$ [3]. These data indicate the necessity of further experiments in order to clarify the situation.

In the present paper we report measurements of K_1 and K_3 on octyl-cyano-biphenyl (OCB). The material, syntetised in our laboratory, has a weakly first order nematic-smectic A transition at 30.9°C ; the nematic-isotrop transition was found to be at 37.6°C .

The elastic constants have been determined by measuring the amplitude of the splay deformation, induced by an electric field in a homogenously aligned sample. In the measurements the field has been kept constant and the temperature scanned. As described in detail in Sec. 2., this method gives a precise determination of the transition temperature, and a check whether the transition is of first or of second order. The measurements provide the anisotropy of the refractive indices also. The results are analysed in Sec. 3.

In Sec. 4. we describe the structures obtained in the smectic phase by cooling down the sample in a deformed state.

2. Experimental method

In the experiments homogenously aligned sandwich-cells have been used (director parallel to the boundaries of the cell). OCB has a large positive dielectric anisotropy, thus the alignment can be deformed by an electric field. The theory of deformation in electric field has been published in [4].

The deformation starts at a threshold voltage

$$V_{Cr} = \pi \sqrt{\frac{K_1}{\epsilon_a}} \quad /2/$$

For small deformations the angle between the director and the boundaries (θ) is

$$\theta = \theta_m \sin \frac{\pi}{L} z \quad (L \text{ sample thickness}) \quad /3/$$

θ_m and the applied voltage, V , are related as

$$\frac{1}{4} \theta_m^2 = \frac{1}{\epsilon_a / \epsilon_{\perp} + K_3 / K_1} \frac{V - V_{cr}}{V_{cr}} \quad /4/$$

In the experiments θ_m has been determined by measuring the

optical phase difference ($\Delta\phi$) between the ordinary and extraordinary components of a laser beam. For small deformations

$$\frac{1}{4} \theta_m^2 = \frac{n_o^2}{n_e(n_e + n_o)} \frac{\Delta\phi_o - \Delta\phi}{\Delta\phi_o} \quad /5/$$

with $\Delta\phi_o = \frac{2\pi}{\lambda} L (n_e - n_o)$

Thus measuring $\Delta\phi$ as a function of V , K_1 and K_3 can be determined. From the measurement of $\Delta\phi_o$, $\Delta n = n_e - n_o$ can be calculated. (In principle n_e and n_o should be known separately for the calculation of K_3 . However the factor $n_o^2 / n_e(n_e + n_o)$ changes little with the change of n_o , and n_o itself changes little with the temperature. In the calculation we have put $n_o = 1.5$.) The dielectric data are taken from [5].

The approximation for small deformations presented above, works well only for $K_3/K_1 \sim 1$. For higher K_3/K_1 ratios (> 3) it is hard to measure in the limit where this approximation is valid. Thus near the transition, where K_3/K_1 increases rapidly, the elastic constants were determined by comparing the measured data with exactly calculated $\Delta\phi(V)$ curves.

In the actual measurements the applied voltage has been fixed and the temperature has been decreased with a rate of

$\sim 0.05^\circ/\text{min}$. The intensity of the laser beam which gives $\Delta\phi$ is recorded on an X-Y recorder as the function of the temperature. A typical plot is given on Fig. 1. As it can be seen, this method gives a precise determination of the transition temperature, T_{SN} . T_{SN} has been reproduced in different runs within 0.02° .

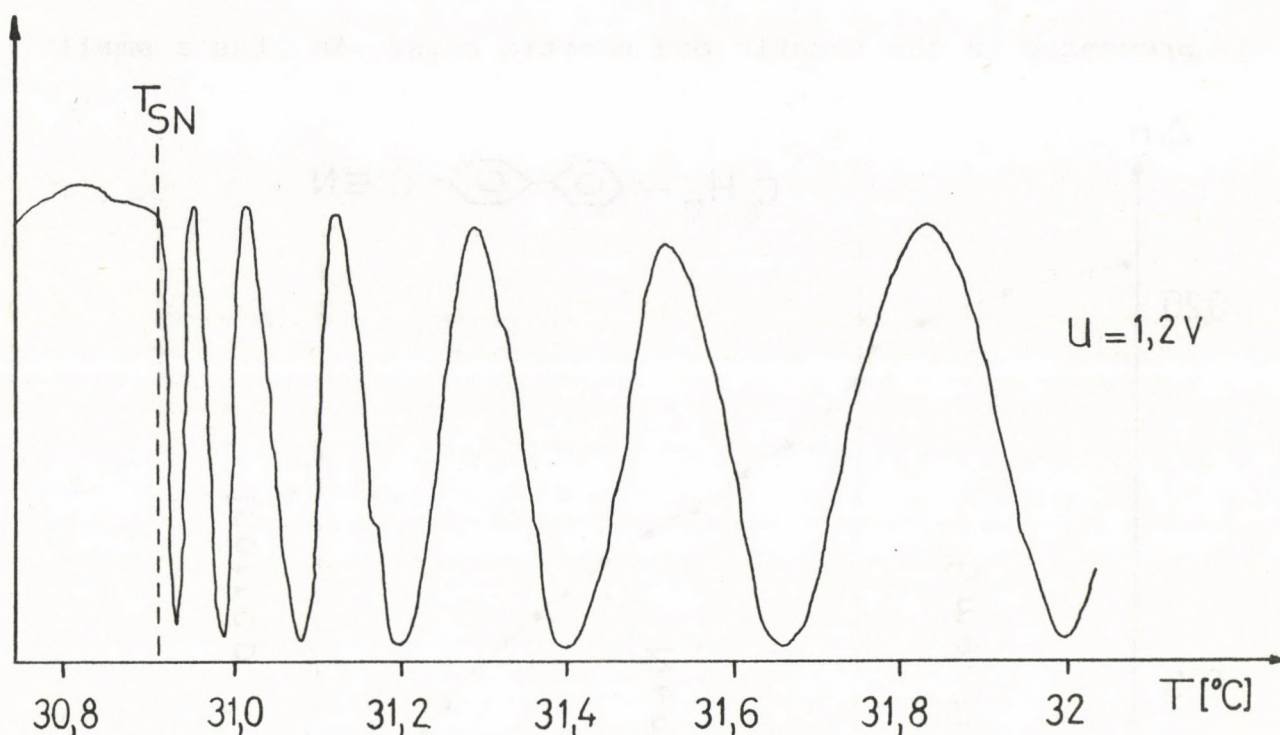


Fig. 1.

Another advantage of this method is that it provides a sensitive check, whether the transition is of second or of first order. At a strictly second order transition K_3 goes to infinity, consequently Θ_m becomes zero, i.e. $\Delta\phi = \Theta\phi_0$. In our measurements a small difference was found between $\Delta\phi$ and $\Delta\phi_0$ at the transition when field have been applied. This fact

clearly demonstrates the first-order character of the transition.

3. Results and discussion

On Fig. 2 the anisotropy of the refractive indices (Δn) is presented in the nematic and smectic phase. Δn has a small

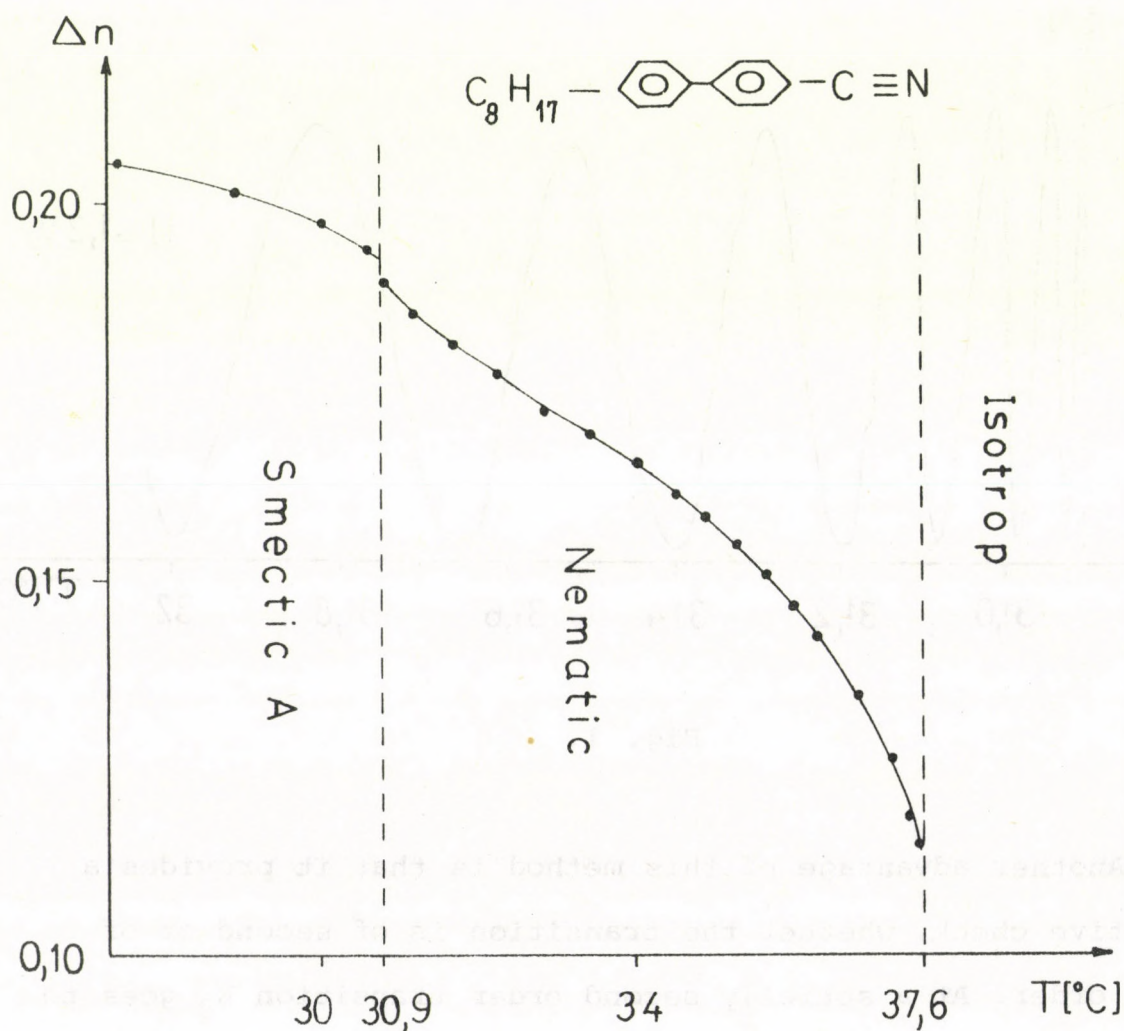


Fig. 2: The anisotropy of the refractive indices as a function of the temperature. $\lambda = 0.63 \mu\text{m}$.

discontinuity ($\sim 1\%$) at T_{SN} , which indicates again that the transition is weakly first order.

The anisotropy of the refractive indices is in good approximation proportional to the orientational order parameter. The shape of the Δn curve presented here agrees qualitatively with McMillan's calculation of the order parameter [9]. However a deviation is found also; the curve has an inflexion at about the same temperature where the pretransitional effects appear. From this fact we conclude that the presence of small smectic regions has an observable influence on the orientational order parameter already in the nematic phase. This influence is neglected in McMillan's theory.

On Fig. 3 the splay elastic constant is presented near the transition temperature. There is a slight increase of K_1 , however, as expected, no divergence has been found.

On Fig. 4 the K_3/K_1 ratio is shown. The pretransitional increase of this ratio starts at about 1° above the transition. Although the transition is of first order, K_3/K_1 becomes very large at the transition. From the $\Delta\phi - \Delta\phi_0$ values, measured at the transition temperature we estimate $K_3/K_1 (T_{SN}) \approx 30$.

To make an analyse of the data in terms of a power-law, we estimate the part of K_3/K_1 , which is due to the nematic or-

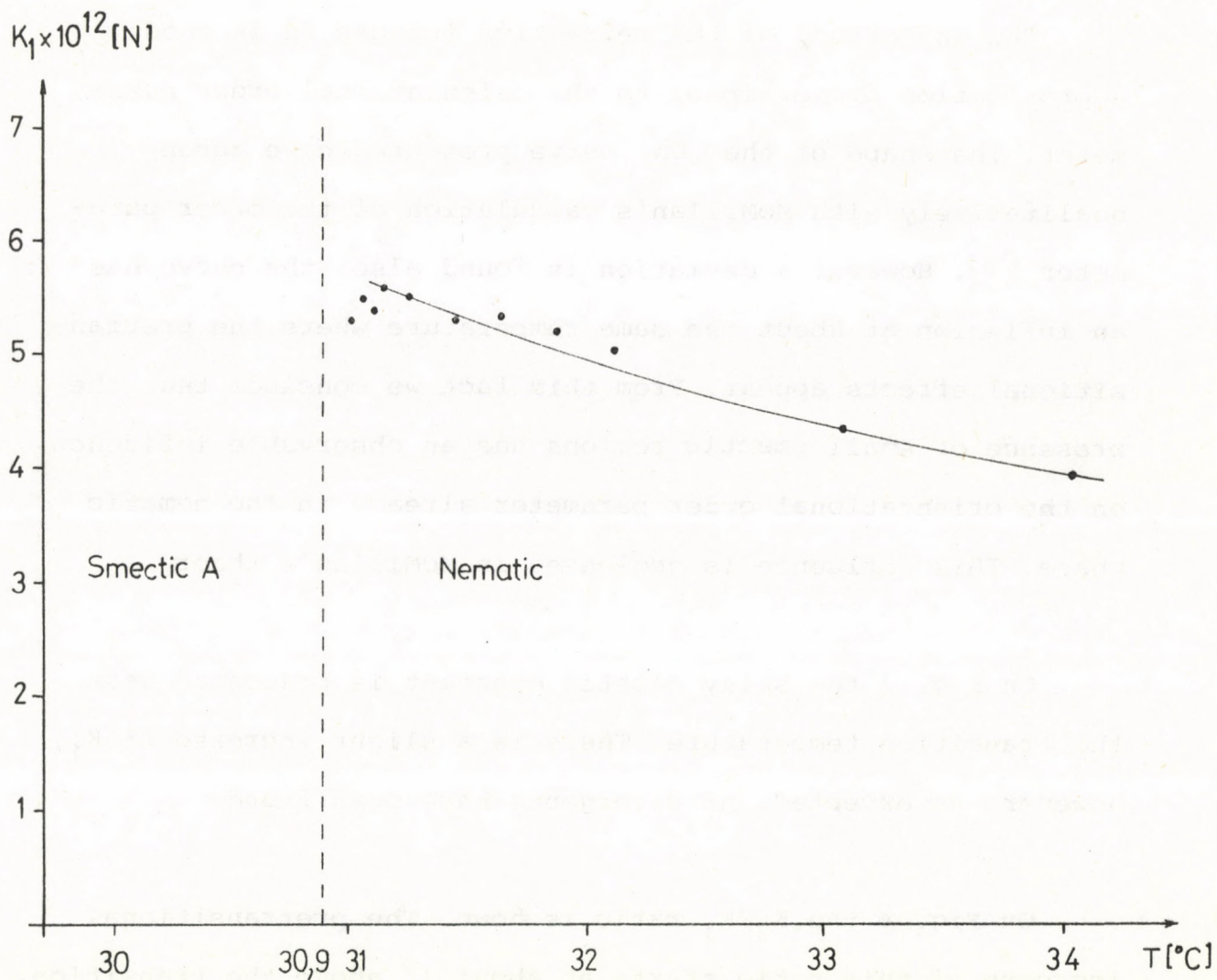


Fig. 3: Variation of the splay elastic constant near the transition

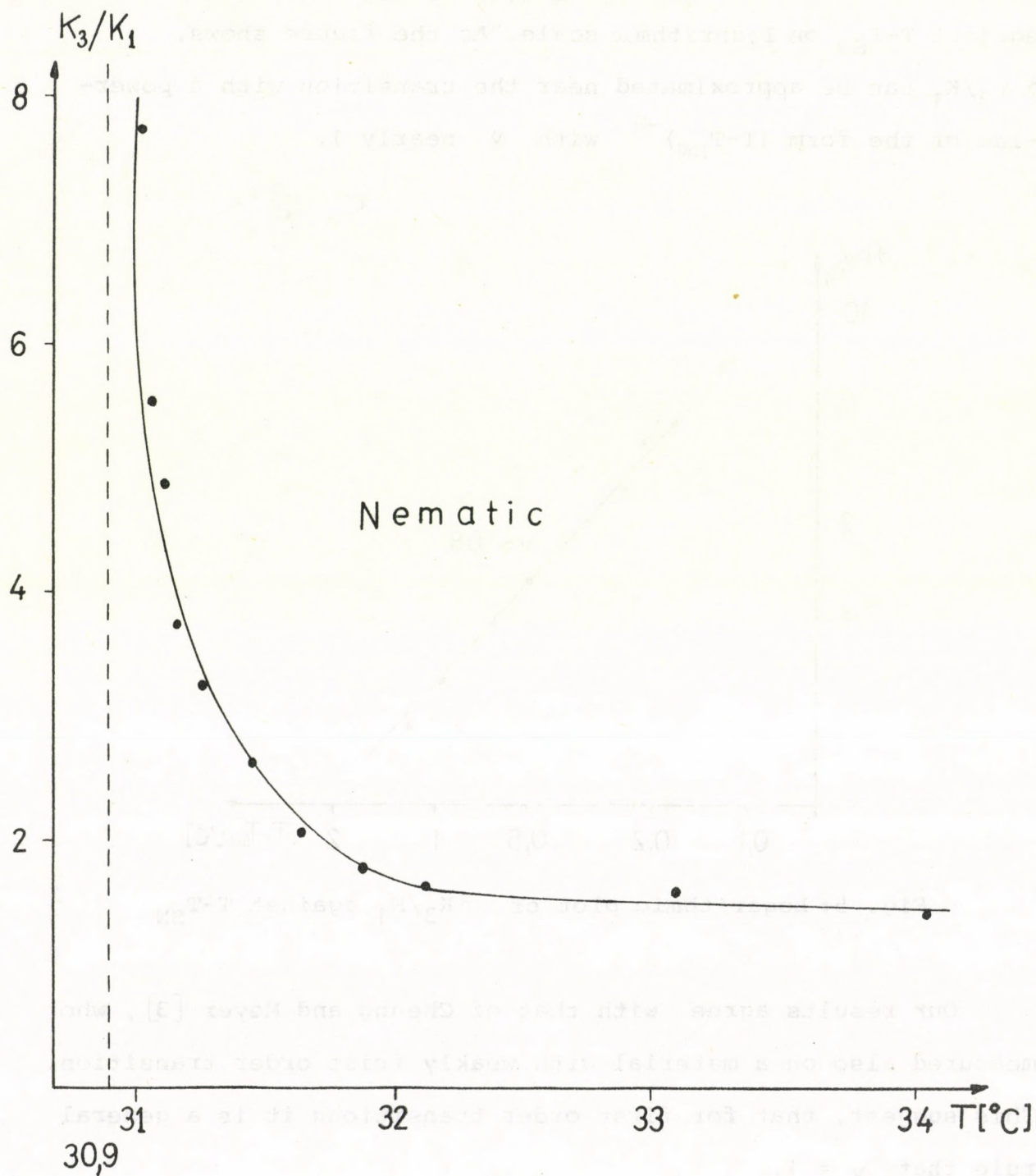


Fig. 4: The K_3/K_1 ratio near the transition

der, to be 1.2. On Fig. 5 $\delta K_3/K_1 = K_3/K_1 - 1.2$ is plotted against $T-T_{SN}$ on logarithmic scale. As the figure shows, $\delta K_3/K_1$ can be approximated near the transition with a power-law of the form $(T-T_{SN})^{-\nu}$ with ν nearly 1.

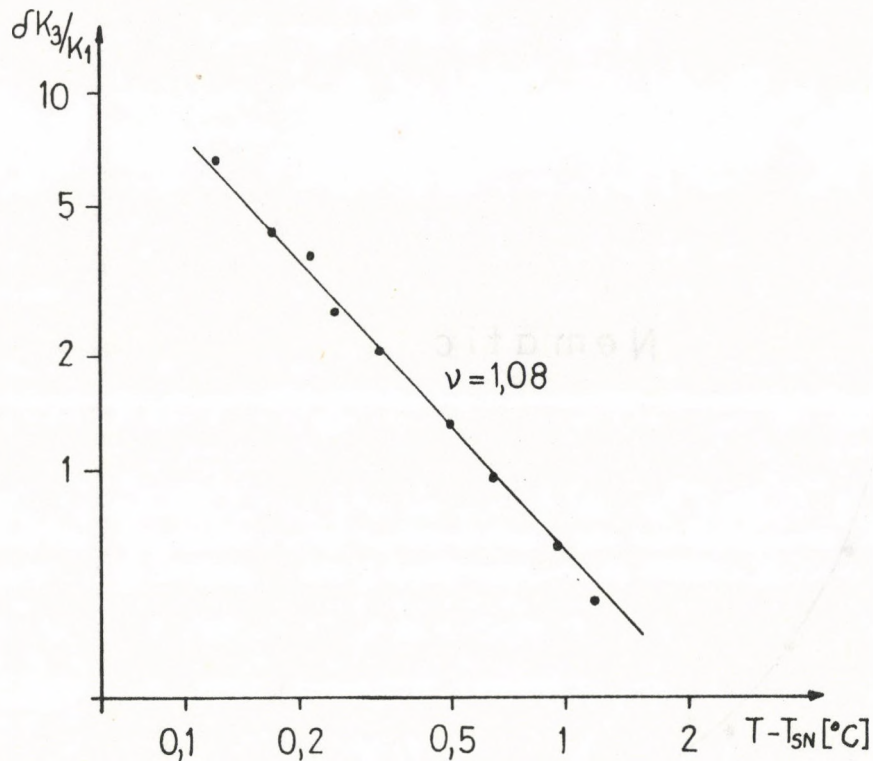


Fig. 5: Logarithmic plot of $\delta K_3/K_1$ against $T-T_{SN}$

Our results agree with that of Cheung and Meyer [3], who measured also on a material with weakly first order transition. This suggests, that for first order transitions it is a general rule that $\nu = 1$.

4. "Frozen in" structure in smectic phase

As mentioned in the previous sections, when the sample is cooled in the presence of a deformation, induced by an electric field, the deformation does not disappear when the transition temperature is reached. At the transition we have

$$\Theta = \Theta_m(V, T_{SN}) \sin \frac{\pi}{L} Z.$$

It has been found, that this deformation freeze in if Θ_m is not too large, i.e. it remains unchanged in the smectic phase even after the field is suppressed. This effect has been deduced from the birefringent measurements; the optical phase difference follows the same temperature dependence as $\Delta\phi_0$, but it is shifted with the value $\Delta\phi_0(T_{SN}) - \Delta\phi(T_{SN})$. It is confirmed by conoscopic observations also; the hiperbola are shifted when the phase transition is passed in a deformed state.

The arrangement of the smectic planes in the undeformed ($\Theta_m = 0$) and deformed ($\Theta_m \neq 0$) states are shown on Fig. 6. The deformation is mainly splay, however a weak bend is also present (proportional to Θ_m^2). Such a structure can be stabilised by formation of edge dislocations [6]. A relaxation to the homogenous orientation is not possible, as it would imply a hydrodynamic motion in which the planes had to split.

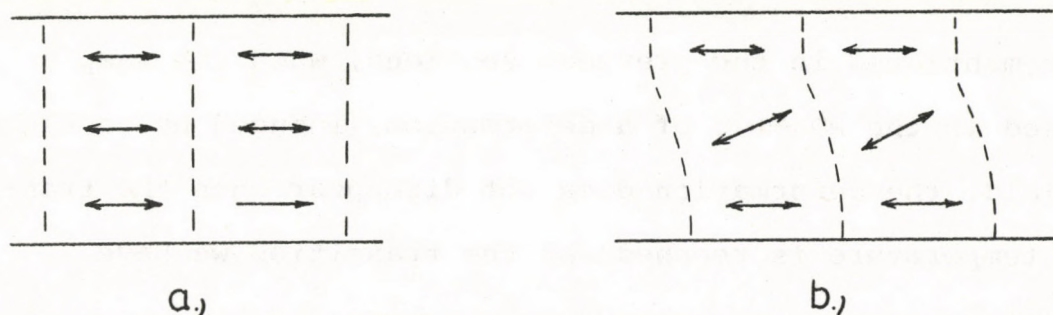


Fig. 6: Undeformed (a./) and deformed (b./) smectic planes. \longleftrightarrow director; - - - planes.

We note that this type of deformation is different from the Parodi-type transition [7]. In the latter case a periodic array of defects is formed, while in our case the deformation is homogenous in the plane of the sample.

As θ_m is increased, a growing number of focal conics are formed in the smectic phase, making impossible birefringent measurements. As observed in polarising microscope, the eccentricity of the focal conics is decreasing as the deformation is increased. This is explained by the relation between the eccentricity of the ellipse (r) and the orientation in the middle of the sample, given in [8]:

$$\text{ctg}^2 \theta_m = r^2 - 1$$

When θ_m is small, r is large (elongated focal conics);

as $\theta_m \rightarrow \frac{\pi}{2}$, $r \rightarrow 1$, i.e. the ellipse becomes a circle.

It has been found that reheating the sample to nematic phase the contours of the focal conics remain visible at the surfaces of the cell. This fact indicates that the formation of focal conics alter slightly the surface orientation also.

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